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Membranes on rough self-affine surfaces

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We study the effective potential U_e between a rough self-affine surface, and a membrane bounded by this surface. We find that the effect of the substrate roughness exponent H on U_e is significant for $\zeta < \xi$. ζ is a healing length, and ξ the in-plane roughness correlation length. However, the effect of H is negligible for membrane-surface separations larger than a characteristic membrane-surface separation τ . Moreover, the roughness contribution to U_e scales as $\sim \zeta^{2H}$ for $\zeta < \xi$, and as $\sim \zeta^{-4}$ for $\zeta \gg \xi$. [S0163-1829(96)08235-5]

I. INTRODUCTION

Interfaces and membranes are topics of significant recent interest in theoretical and experimental physics.¹ Usually, an interface represents a boundary between two phases whose fluctuations can be studied by methods employed in equilibrium critical phenomena. It is formed from the same molecules that constitute the bulk phases, and has a limited internal structure. Moreover, surface tension ensures that such surfaces remain relatively flat.¹ In contrast, membranes are composed of molecules different than the medium in which they are imbedded, and do not necessarily separate two distinct phases. They have significant internal structure, entailing rigidity, ordering of various sorts, etc. Furthermore, since their surface tension vanishes or is small, membranes can exhibit wild surface fluctuations.¹ Thermally induced fluctuations on membranes around their lowest-energy conformation can produce long-range steric repulsion² between two membranes, or a membrane and a substrate. The competition between these repulsive forces and direct attractive forces, i.e., van der Waals forces³ could result in an unbinding transition for which experimental evidence has already been found.⁴

Interface/surface fluctuations give rise to fundamental contributions in a variety of processes,⁵⁻⁷ and systems with membranes.^{2-4,8-10} Extensive studies have been performed already for membranes bounded by flat and uniform substrates.^{1-4,8-10} However, real substrate surfaces are always characterized by some degree of roughness that depends on the material, the method of surface treatment, and the presence of absorbed species. Recently, the role of roughness on wetting interface phenomena has attracted the attention of several authors.^{11,12} Nevertheless, an extension of these studies to membranes is still in its infancy or missing.

Therefore, we will study the case of membranes completely or partially bounded by a rough surface in what is usually referred as the interface approach theory.¹¹ In terms of this approach, the Hamiltonian $H[h/z] = \int (K/2)(\nabla^b h)^2 + U[h(r) - z(r)]d^2r$ describes with surface tension K for $b=1$, and membranes with bending rigidity K for $b=2$. The membrane/interface profile is denoted by

$h(r)$, the substrate height profile by $z(r)$, and the interface/membrane substrate interaction potential by $U[h(r) - z(r)]$ with $r=(x,y)$ the in-plane position vector. We recall that in the case of a flat substrate $z(r)$ equals zero.

In general, the study of the membrane asymptotic properties due to substrate roughness^{11,12} (similarly to interfaces) requires a specific local and global characterization of the substrate roughness. The roughness causes a deformation of the membrane to a degree that depends on the bending rigidity, and the substrate interatomic potential. Moreover, the effect of substrate height fluctuations will appear as a correction to the leading scaling behavior of the interaction potential. In our paper, we will examine the roughness effect for membranes bounded on self-affine rough surfaces, under a general scheme that applies to this category of rough surfaces.

II. THEORY FOR MEMBRANES

In the more general case, membranes are characterized not only by the bending rigidity K but also by a "lateral tension" R , which plays a similar role to the surface tension for an interface.¹³ Lateral tension can suppress membrane fluctuations, and can alter significantly the adhesive properties of membranes.¹⁴ For any finite tension, a planar membrane should behave at large length scales like a fluid interface that is bound for any long-range attractive forces.¹⁵ On the other hand, for closed membranes, i.e., vesicles, lateral tension can arise from the constraints of a fixed surface area and enclosed volume,¹⁶ in agreement with results from micropipet experiments.¹⁷

In the interface potential approach theory,¹¹ the membrane is attached to the substrate and forms a layer close to it. The regime of validity of this approach is confined to substrate and layer fluctuations such that $h(r) - z(r)$ is much larger than the bulk correlation length of the membrane layer.^{11,12} The system is described by the Hamiltonian

$$H[h,z] = \int \frac{K}{2}(\nabla^2 h)^2 + \frac{R}{2}(\nabla h)^2 + U[h(r) - z(r)]d^2r, \quad (1)$$

which captures the correct scaling behavior in the limit of large membrane-substrate separations. The interaction potential $U[h(r)-z(r)]$ results from the addition of two-body substrate-membrane interactions, and in general is a nonlocal function of $h(r)$ and $z(r)$.

The membrane surface profile is obtained (under the restriction of absence of thermal fluctuations) by the minimization of Eq. (1), which yields the Euler-Lagrange equation,

$$U'(h-z) - R\nabla^2 h(r) + K(\nabla^2)^2 h(r) = 0. \quad (2)$$

Expansion of $U(h-z)$ around a minimum value “ w ” [$U'(w)=0$] yields $U(h-z) \approx U(w) - (1/2)U''(w)(h-z-w)^2$. Its substitution in Eq. (2) finally gives

$$U''(w)(h-z-w) - R\nabla^2 h(r) + K(\nabla^2)^2 h(r) = 0, \quad (3)$$

since $U'(h-z) = U''(w)(h-z-w)$. Fourier transformation of Eq. (3) yields

$$h(q) = \frac{U''(w)}{U''(w) + Rq^2 + Kq^4} z(q) + w\delta(q). \quad (4)$$

In comparison to the case of interfaces ($K=0$),^{11,12} Eq. (4) is the more general expression that relates interface/membrane and substrate height fluctuations. In deriving Eq. (4), the definition $[h,z](r) = \int [h,z](q) e^{-iqr} d^2q$ was used for the Fourier transforms.

The “healing” length yields the maximum wavelength of the undulations of the solid-substrate surface that are reproduced by the membrane surface. From Eq. (4) we can distinguish two characteristic length scales. One is the “healing” length $\zeta = [K/U''(w)]^{1/4}$ of the pure membrane problem ($R=0, K \neq 0$), and the other the “healing” length $Y = [R/U''(w)]^{1/2}$ of the pure “wetting-interface” problem (with R playing the role of surface tension).^{11,12} The ratio ζ^2/Y separates two regimes of length scales (L) such that for $L < \zeta^2/Y$ the bending rigidity dominates, while for $L > \zeta^2/Y$ the lateral tension contribution is the dominant one. This is obvious, if we compare the terms $\zeta^4 q^4$ and $Y^2 q^2$ from the denominator of Eq. (4) with $q=1/L$. In addition, there will be a lateral tension $R^* = [KU''(w)]^{1/2}$ ($Y=\zeta$) such that computations valid for $R=0$, will also apply for $R \ll R^*$.

From Eq. (4), we can determine the healing length S in the case where $R \neq 0$ and $K \neq 0$, if we consider the equality $Rq^2 + Kq^4 = U''(w)$ with $q=1/S$.⁹ Solving for S , assuming $K \neq 0$, leads to $S^{-2} = [(4\zeta^4 + Y^4)^{1/2} - Y^2]/2\zeta^4$. For $Y \gg \zeta$, we obtain $S^{-2} \approx Y^{-2}$ while for $Y \ll \zeta$, $S^{-2} \approx \zeta^{-2}$. If the roughness wavelength ξ is larger than the healing length S , the membrane follows the roughness of the substrate while curvature rigidity and lateral tension plays a negligible role. In the opposite case $|h(q)|$ is small, and the membrane surface remains “flat.”

Fourier transformation of Eq. (1), substitution from Eq. (4), and ensemble average over possible roughness realizations yield the effective potential $U_e(w) = \langle H[h,z] \rangle$,

$$U_e(w) = AU(w) + F(\zeta, Y) \quad (5)$$

with

$$F(\zeta, Y) = \frac{(2\pi)^2}{2} \int \frac{\zeta^4 q^4 + Y^2 q^2}{1 + \zeta^4 q^4 + Y^2 q^2} \langle |z(q)|^2 \rangle d^2q, \quad (6)$$

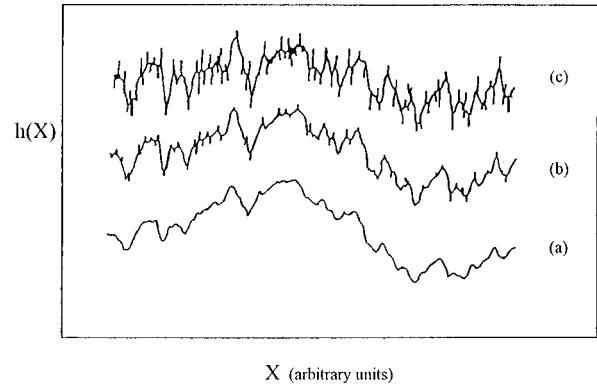


FIG. 1. Schematics of the surface height profile $h(X)$ vs X for various values of the roughness exponent H : (a) $H=0.8$, (b) $H=0.5$, (c) $H=0.2$. The corresponding local fractal dimension is $D=3-H$ for two-dimensional substrates, and $D=2-H$ for one-dimensional substrates.

where the factor A denotes the macroscopic average flat surface area. In Eq. (6), the terms $Kq^4|z(q)|^2$ and $Rq^2|z(q)|^2$ represent, respectively, the lateral tension and bending rigidity energy cost if the membrane follows the substrate fluctuations at a wave vector q . The term $U''(w)|z(q)|^2$ represents the substrate interaction cost if the membrane profile becomes flat. The term $F(\zeta, Y)$ in Eqs. (5) and (6) represents the energy cost to deform the membrane due to substrate roughness. The integration in Eq. (6) is in the range $0 \leq q \leq Q_c (= \pi/a_0)$ with a_0 the atomic spacing. Figure 2 shows the roughness contribution $F(\zeta, Y)$ to $U_e(w)$ for various values of Y relative to ζ and S^{-2} (inset).

For sufficiently large lateral tension ($R \gg R^*$), we have $Y \gg \zeta$, which results in $Y^{-2} \approx S^{-2}$. Furthermore, in this case we can neglect the curvature effect ($K \approx 0$) resulting in the asymptotic behavior $F \sim Y^{-2}$ at $Y \gg \xi$ as is observed in wetting of fluid interfaces.¹² Subsequently, large lateral tension yields $F \sim S^{-2}$ in the asymptotic regime $S(\approx Y) \gg \xi$. Indeed, such a behavior is in agreement with the fact that at large length scales a planar membrane (with finite lateral tension) should behave like a fluid interface.¹⁵

III. SELF-AFFINE FRACTAL ROUGHNESS

All rough surfaces exhibit perpendicular fluctuations that are characterized by a root mean-square deviation from flatness $\sigma = \langle z(r)^2 \rangle^{1/2} [\langle z(r) \rangle = 0]$ with $\langle \dots \rangle$ an average over the whole planar reference surface. For an isotropic random surface, the height-height correlation function is written as $C(r) = \langle z(0)z(r) \rangle$ where the average is taken over all pairs of points on the surface, which are separated by a distance $|r|$. The correlation function $C(r)$ for any physical self-affine surface¹⁸ is characterized by a finite correlation length ξ (which is a measure of the average distance between peaks and valleys on the surface) such that $C(r) \approx \sigma^2 - Dr^{2H}$ for $r \leq \xi$, and $C(r) = 0$ for $r \gg \xi$ (with $D \sim \sigma^2/\xi^{2H}$ is a constant). The roughness exponent $0 < H < 1$ is a measure of the degree of surface irregularity.^{18,19} Small values of H (~ 0) characterize extremely jagged or irregular surfaces, while large values H (~ 1) surfaces with smooth hills and valleys.^{18,19} For an illustration of the effect of H see Fig. 1.

The correlation function $C(r)$ is the Fourier transform of $\langle |z(q)|^2 \rangle$ and vice versa. An analytic correlation model in Fourier space (k -correlation model) for self-affine structures was given already in earlier studies.²⁰ This model is valid for the whole range of values for the roughness exponent $0 \leq H < 1$. Following the notation of Ref. 20, $\langle |z(q)|^2 \rangle$ is given

$$\langle |z(q)|^2 \rangle = \frac{A}{(2\pi)^5} \frac{\sigma^2 \xi^2}{(1 + a q^2 \xi^2)^{1+H}}, \quad (7)$$

where the normalization condition

$$[(2\pi)^2/A] \int_0 < q < Q_e \langle |z(q)|^2 \rangle d^2 q = \sigma^2$$

yields the parameter “ a ” after substitution of Eq. (7):

$$a = \frac{1}{2H} [1 - (1 + a Q c^2 \xi^2)^{-H}] \quad (0 < H < 1), \quad (8)$$

$$a = \frac{1}{2} \ln(1 + a Q c^2 \xi^2) \quad (H = 0).$$

Expressions valid for $H=0$ can be obtained from those valid for $H>0$, if we consider the identity $\lim_{H \rightarrow 0} (1/H)[x^H - 1] = \ln(x)$. The limiting case of logarithmic roughness ($H=0$) is related to predictions of various growth models for the nonequilibrium analogue²¹ of the equilibrium roughening transition.²²

IV. ANALYTIC AND NUMERICAL RESULTS

A. Long-range interaction limit

We will limit our discussion to the case of zero lateral tension ($R=0$) and nonzero bending rigidity ($K \neq 0$) with $F(\zeta) = F(\zeta, Y=0)$, since fluid membranes are dominated by weak curvature elasticity.^{1,13} If we consider the case of small healing lengths $\zeta \ll \xi$, where we omit the regime of small wave vectors q from the roughness spectrum or $\langle |z(q)|^2 \rangle = [A \sigma^2 \xi^2 / (2\pi)^5] a^{-1-H} (q \xi)^{-2-2H}$, we obtain from Eq. (6)

$$U_e(w) \approx A U(w) + G(H) U''(w) \zeta^{2H}, \quad (9)$$

$$G(H) \approx [\sigma^2 / (4 - 2H) (2\pi)^6 a^{1+H} \xi^{2H}] \int (1 + x^2) x^{1-H} dx, \quad (10)$$

where a power-law behavior similar to Eq. (9) was also observed for wetting of interfaces; $U_e(w) - U(w) \sim Y^{2H}$.^{11,12}

If we consider long-range³ substrate-membrane interactions $U(w) \approx u w^{-(s+1)} + c w^{-s} \dots$ (u, c are constants), we obtain $U_e(w) \approx U(w) + \delta U(w)$ with $\delta U(w) \sim w^{-(s+1)(1-H/2)}$ as the leading correction to the potential U_e due to bending rigidity. For consistency, the correction term $\delta U(w)$ must be weaker than the original potential $U(w) \sim w^{-(s+1)}$. As a result we obtain $s - 1 < (s+1)(1 - H/2)$ or $H < 4/s + 1$. Following the terminology of Ref. 23, we can distinguish a “mean-field” regime ($H < 4/s + 1$) where the asymptotic behavior is dominated by the original potential $U(w)$ and substrate roughness is irrelevant, and a “strong-fluctuation” regime ($H > 4/s + 1$) where the bending rigidity energy dominates the original potential $U(w)$. In

the strong-fluctuation regime, the interaction potential $U_e(w)$ has to be obtained in a self-consistent manner or $U_e(w) \sim U e''(w)^{(1-H/2)}$, which yields $U_e(w) \sim w^{4(1-H/2)/H}$.¹¹

For van der Waals forces or $s=3$,³ we have, respectively, $H < 4/3$ and $H < 1/2$ for the mean-field regimes of membranes ($K \neq 0, R=0, H < 4/s + 1$) and wetting of interfaces ($R \neq 0, K=0, H < 2/s + 1$).^{6,7} Thus, for membranes the mean-field regime is extended to the whole range of values of the roughness exponent $0 < H < 1$, which can be important experimentally. More precisely, in quartz crystal microbalance (QCM) roughness studies,²⁴ the determination of the roughness exponent H for $s=3$ was feasible for wetting of interfaces in the regime $H < 1/2$. However, QCM studies with membranes seem to be a more promising tool for roughness studies since the mean-field regime covers the whole range $0 \leq H < 1$.

B. Logarithmic roughness ($H=0$)

For $H=0$, we can determine $F(\zeta)$ exactly since in this case the associated integral has an analytical form. After performing the integration of Eq. (6) we obtain

$$F(\zeta) = A U''(w) \frac{\sigma^2 \xi^2}{16\pi^2} (X_1 + X_2) \zeta^{-2}, \quad (11)$$

where

$$X_1 = -\zeta^{-4} (\zeta^4 + a^2 \xi^4)^{-1} \tan(\zeta^2 Q_c^2) + a \zeta^2 \xi^2 [2(\zeta^2 + a^2 \xi^4)]^{-1} \ln(1 + \zeta^2 Q_c^2),$$

and $X_2 = \zeta^6 [a \xi^2 (\zeta^4 + a^2 \xi^4)^{-1}] \ln(1 + a \xi^2 Q_c^2)$. In the limits $\zeta \ll \xi$ and $\zeta \gg \xi$, Eq. (11) yields, respectively,

$$F(\zeta) = A \frac{K \sigma^2}{32\pi^2} \ln[a Q c^2 \xi^2] \zeta^{-4} \quad (\zeta \gg \xi), \quad (12)$$

$$F(\zeta) = A \frac{K \sigma^2}{32\pi^2} \ln[Q c^2 \xi^2] \zeta^{-4} \quad (\zeta \ll \xi). \quad (13)$$

The limiting behavior given by Eq. (12) will be important to determine the asymptotic behavior in the regime $\zeta \gg \xi$ also for $H > 0$.

C. Self-affine roughness ($0 < H < 1$)

Further calculations of $F(\zeta)$ for values of the roughness exponent H in the range $0 < H < 1$ can only be performed numerically. This can be seen in Fig. 3 where we plot the roughness term $F(\zeta)$ versus ζ for $H=0, 0.5, 0.9$ and $\xi=60$ nm. The choice for this value of ξ is based on observations for this size of correlation lengths for metallic (Ag or Au) films in experimental roughness studies.¹⁸ Figure 3 shows clearly that for $\zeta < \xi$ surfaces with various degrees of irregularity can be clearly distinguished. Moreover, the roughness contribution as a function of H becomes maximum for the case of logarithmic roughness ($H=0$). Thus, substrate fluctuations that resemble those of capillary waves in liquids at $H=0$ affect more drastically the free membrane profile.

For large healing lengths $\zeta \gg \xi$, all curves in Fig. 3 merge together, resulting in loss of any memory from the substrate

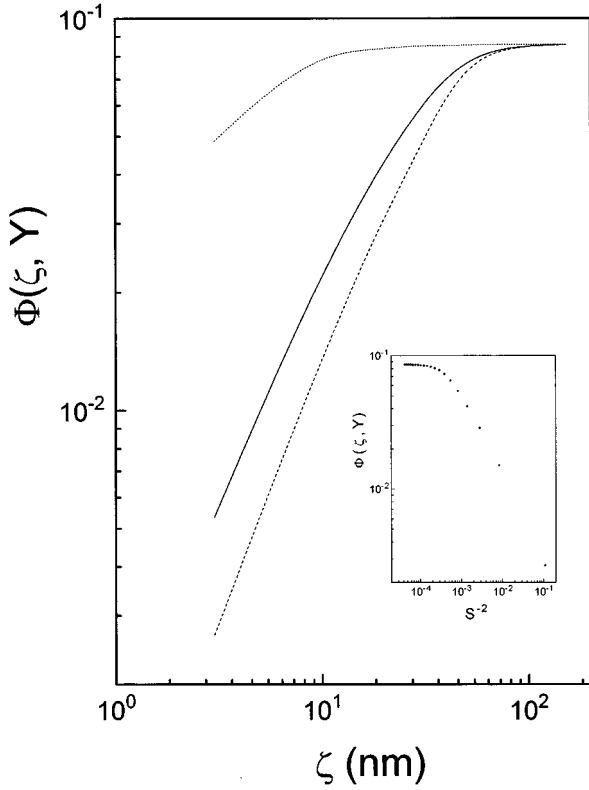


FIG. 2. Log-log plots of $\Phi(\zeta, Y) = F(\zeta, Y)/U''(w)A$ from Eqs. (6) and (7) as a function of ζ for $H=0.7$, $\xi=60$ nm, $\sigma=3$ nm, $a_0=0.3$ nm. Dots, $Y=0.1\zeta$; solid line, $Y=\zeta$; dashes, $Y=10\zeta$. The inset shows $\Phi(\zeta, Y)$ as a function of S^{-2} for $H=0.7$, $\xi=60$ nm, $\sigma=3$ nm, $a_0=0.3$ nm, and $Y=0.1\zeta$.

fluctuation density as is described by the roughness exponent H . Moreover, we can calculate the effective potential $U_e(w)$ for all values of H since for $H=0$ such a calculation is possible [Eq. (12)], and from Fig. 3 all curves merge together each. As a result the asymptotic behavior is the same at large healing lengths. Thus, we can conjecture the general asymptotic behavior of $U_e(w)$ for all $H(0 \leq H < 1)$ and $\zeta \gg \xi$:

$$U_e(w)/A \approx U(w) + \Omega \zeta^{-4}, \quad \Omega \approx \frac{K \sigma^2}{32 \pi^2} \ln[a Q_c^2 \xi^2]. \quad (14)$$

In the corresponding case of a wetting interface ($K=0, R \neq 0$), we had the asymptotic behavior $U_e(w) - U(w) \sim Y^{-2}$.¹²

For substrate in-plane correlation length ξ of the same size as the healing length ζ , we obtain a crossover from the regime where the free membrane interface is controlled to a significant degree by substrate fluctuations to a regime where it is unaffected. Thus, we can estimate the effective membrane-surface separation τ that determines such a behavior for a substrate with a given correlation length ξ . If we consider long-range interactions for the flat substrate $U(w) \sim w^{1-s}$ [$U''(w) \sim s(s-1)w^{-1-s}$], we obtain after substitution in $\xi = \zeta (= [K/U''(w)]^{1/4})$ the critical membrane-surface separation,

$$\tau \sim [D(s-1)s/K]^{1/s+1} \xi^{4/s+1}. \quad (15)$$

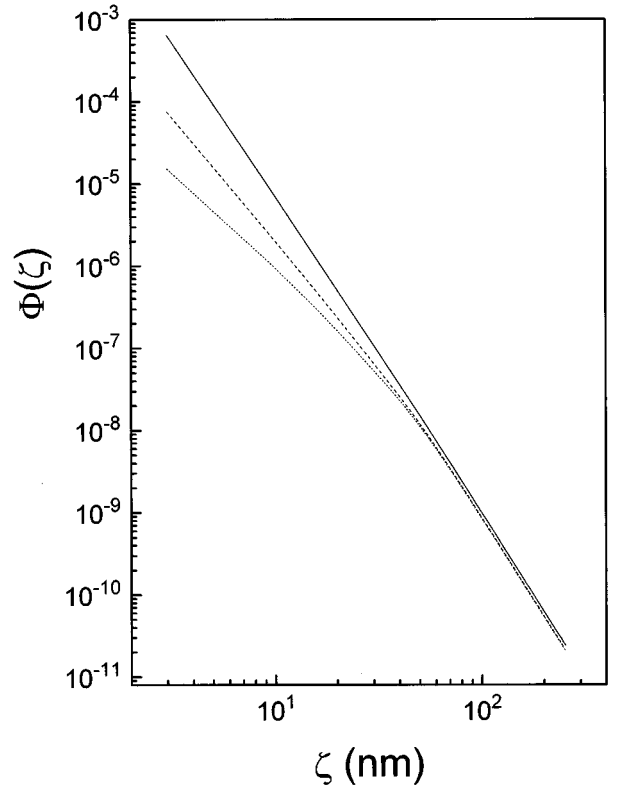


FIG. 3. Log-log plots of $\Phi(\zeta) = F(\zeta, Y=0)/KA(R=0)$ from Eqs. (6) and (7) for $\sigma=3$ nm, $\xi=60$ nm, $a_0=0.3$ nm as a function of the healing length ζ . Solid line, $H=0$; dashes, $H=0.5$; dots, $H=0.9$.

In Eq. (15), the exponent $4/s+1$ is the one that controls the crossover from the mean-field regime ($H < 4/s+1$), to the strong-fluctuation regime ($H > 4/s+1$). For the case of van der Waals forces ($s=3$), the critical membrane-surface separation τ scales as function of ξ as $\tau \sim \xi$. For the wetting problem of interfaces, we had $\tau \sim \xi^{2/s+1}$ and $\tau \sim \xi^{1/2}$ for $s=3$.^{11,12}

V. CONCLUSIONS

In conclusion, we investigated the effect of self-affine and logarithmic surface roughness on the effective potential for membranes bounded by rough surfaces. Such a study became feasible by combining membrane theories for rough surfaces with that of analytic surface self-affine correlation models. We emphasized more the case of fluid membranes that are dominated by weak curvature elasticity¹ with zero lateral tension. Nevertheless, our results will be also valid for weak lateral tension ($R \ll R^*$ or slightly stretched membranes) as was pointed out earlier.¹³ Moreover, finite-membrane-size effects can be ignored for roughness correlation lengths ξ (typically $\xi < 100$ nm as observed in many real systems¹⁸) sufficiently smaller than the membrane size of typical magnitude $\sim 1 \mu\text{m}$.^{10,13}

It is found that in order for the substrate roughness to have a significant contribution on fluctuations of the membrane profile (effect of H), the substrate correlation length ξ should be larger than the healing length ζ . This is also the expected result if ones makes a comparison with conclusions

regarding wetting of fluid interfaces.^{11,12} Alternatively, the roughness effect will contribute significantly for membrane layers of membrane-surface separation smaller than a critical membrane-surface separation τ [Eq. (15)]. Finally, for large healing lengths $\xi \gg \xi$, it is shown that the roughness effect scales as $F(\xi) \sim \xi^{-4}$ for values of H in the range $0 \leq H < 1$.

Experimentally, an investigation of the membrane fluctuations induced by the roughness of the substrate on which they are bounded might be possible, because of a recent ingenious experimental development by Radler *et al.*¹⁰ In fact, they combined phase contrast microscopy with reflection in-

terference microscopy,¹⁰ which made it possible to measure both the microscopic shape and the fluctuations of the bound part at the same membrane (vesicle in the particular case).

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